Novel APPJ-thermal hybrid system for methane decomposition

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Significance and Relevance

The combination of an Atmospheric Pressure Plasma Jet (APPJ) with external heating represents a novel hybrid system for decomposing (bio)methane into hydrogen and carbon. As methane splitting occurs at high temperatures even with a catalyst (≥900°C), using cold plasma could be of significant relevance for the activation of CH_4 at room temperature. Consequently, a lower temperature (≤ 750°C) would be required in the subsequent thermal step, thereby facilitating the development of a novel CO_2-free hydrogen production process that is more commercially viable.

Introduction and Motivations

A low-carbon hydrogen production is one of the hot topics of industrial research thank to its several use from chemicals production to energy vector[1], and its use will reach 150 Mt by 2030 making it a high demand commodity.[2]

Decomposition of (bio)methane is an attractive route among low-carbon hydrogen production technologies because it requires approximately 87% lower energy than water electrolysis, but its kinetic needs high temperature, making this process energy extensive. Moreover, inevitable amorphous carbon produced, and soot deposition decreases catalyst activity by time and requires post-reaction separation and purification steps, increasing the overall cost of the process. To cope with the limitations mentioned, in this work a plasma and thermal hybrid system is proposed as financially viable solutions on an industrial scale.

Materials and Methods

The APPJ reactor consists of an inner metallic pipeline acting as AC high-voltage electrode and a coaxially placed quartz tube. Three different configurations (figure 1) without (A) or with ground electrode were tested, consisting of a simple clamp (B), or aluminum foil (C) wrapped around a quartz tube. In the second stage, activated CH_4 flows inside the furnace, where bulk reaction temperature was controlled from 600°C to 750°C. The distance between plasma and thermal zones varied in order to study the effect on conversion and selectivity through hydrogen, carbon, and C2-C3 impurities. For the same reason carbon-based catalysts, commonly used for only thermal decomposition of methane, were placed inside the thermal stage. Optical emission spectroscopy (OES) was used to analyze species formed within the plasma discharge and to determine the physical-chemical parameter as the level of electronic temperature and concentration of activated species.

Results and Discussion

At a fixed plasma power of 20W and a distance of 14.5cm between the two stages, the conversion rate was raised from 1.5% to 3.8% when plasma was combined with the thermal stage in configuration A at a temperature of 600°C. Similarly, results were witnessed in the case of configurations B and C, although there was a less marked increase. A further conversion of 12.8% was observed when there was no distance between the two stages, using configuration A. Moreover, it was demonstrated that no conversion occurred when only the thermal stage was used at the same temperature, which serves to substantiate the beneficial effect of coupling APPJ and thermal methane decomposition. The observed increase in conversion can be attributed to the activation of CH_4 molecules by the energetic plasma environment in the first stage, which provides abundant CH_x-based radicals, as sustained by the plasma peaks determined by OES analysis.[3] Additionally, the short lifetime of active CH_4 explains the increased conversion when there is no distance between the two stages, allowing for a higher decomposition when plasma is generated directly inside the thermal zone. The presence of a high concentration of H radicals also explains the highest selectivity to hydrogen (~40%) across all three configurations, with ethane, solid carbon, ethylene, and a minor amount of propane and propylene as the subsequent products. These findings contribute to a deeper understanding of the plasma thermal hybrid system and provide a comprehensive account of its behavior.

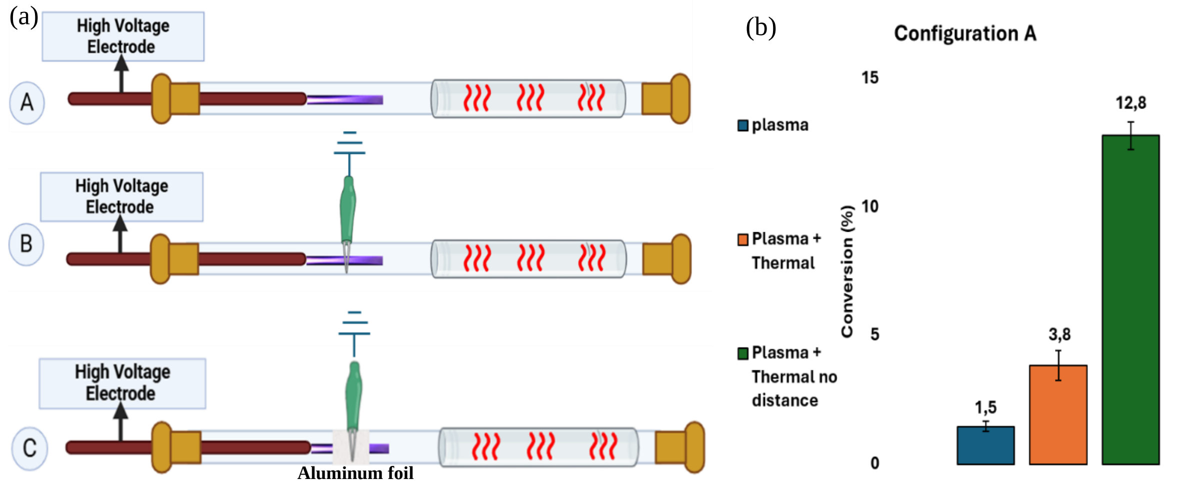

Figure 1 (a) APPJ configurations without (A), or with clamp (B) and aluminum foil (C) as a ground electrode and (b) CH_4 conversion in configuration A.

References

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